

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Examiner: Examiner: Randy BOYER Group art unit: 1797  
Re: Application of: Helmuth SCHULZE-TRAUTMANN, *et al.*  
Serial No.: 10/516,672  
Filed: August 8, 2005  
For: **MICROCRYSTALLINE PARAFFIN, METHOD  
FOR PRODUCING MICROCRYSTALLINE  
PARAFFINS AND USE OF THE  
MICROCRYSTALLINE PARAFFINS**

Confirmation No. 5286

**DECLARATION OF  
Frank Bauer UNDER 37 C.F.R. 1.132**

March 30, 2010

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

I, Frank Bauer, declare and state:

1. I presently hold the position of senior scientist at Leibniz-Institut für Oberflächen-modifizierung e.V., located at Leipzig (Germany).
2. My expert opinion and conclusions, as set forth in this Declaration, are based upon my familiarity with the invention taught by the above-identified patent application (hereinafter "Schulze-Trautmann") and on my study and analysis of the art cited by the Examiner, together with my expertise and experience in the field of heterogeneous catalysis, as evidenced by my about 100 scientific publications (~600 citations within Web of Science), 2 patents and 35 years of research, as well as my experience in the academic and industrial arts relating to chemical engineering, including the processing of Fischer-Tropsch products. My further professional experience and publications are

summarized by my Curriculum Vitae, which is appended to and made a part of this Declaration as Exhibit 1.

3. I have been provided with a copy of the above-mentioned USSN 10/516,672 patent application and a copy of the amended claims included with the Response to the outstanding official action. I have read and understood each of these documents.

4. I have read and understood the Examiner's October 1, 2009 Office Action, which rejects all of the pending claims. In particular, I understand that the Examiner has taken the position that 1) the invention described in the pending claims would have been anticipated by and/or obvious to one of ordinary skill in the art at the time the invention was made in view of Hoek (US2004/0199040) or in view of a combination of Hoek taken in view of Eilers (EP 668342 A1) and/or Bertaux (EP776959 A2). The basis of these rejections is detailed at items 8-21 of the Office Action. In addition, the Examiner has taken the position that 2) the pending claims would have been obvious over Wittenbrink taken in view of Hoek and Bertaux or Carati (US 5,981,419). The basis of these rejections is detailed at items 22-37 of the Office Action. I have also read these references and understand their respective disclosures.

5. The invention is an improved microcrystalline paraffin (for example, claim 25) produced by a new process (for example, claim 31). The Examiner has taken the position that Hoek (US2004/0199040) anticipates or renders obvious the microcrystalline paraffin and/or the process of producing the paraffin.

6. I believe that I am well qualified as an expert in the field to analyze these documents and render an opinion as to what one skilled in the art would be taught by the prior art cited against this patent application.

7. In my opinion, the inventive microcrystalline paraffin and method for producing this paraffin would not have been obvious in view of the information contained in either combination relied upon by the Examiner. My opinion is based upon my extensive

experience as a researcher. The reasons for my opinion are set forth in the following paragraphs.

8. The prior art of processes for the hydroisomerization of long chain n-paraffins by bifunctional catalysts indeed discloses the application of any zeolite for the support of the hydrogenating metal component, but there is no teaching or requirement that the zeolite crystals do not contain any hydrogenation metal component (Schulze-Trautmann, paragraphs 0047 and 0055).

9. The prior art of processes for the hydroisomerization of long chain n-paraffins by bifunctional catalysts indeed claims the reduction of the content of n-paraffins, the minimization of the amount of hydrocracking, and the maximization of the yield of iso-paraffins, but there is no teaching or requirement on the composition of the hydroisomerized waxes (carbon number distribution, iso-to-n paraffin ration, single- and multibranched isomers). Using the zeolite catalyst as described in the claims and the specification of Schulze-Trautmann, the hydroisomerized wax (having reduced contents of hydrocracked C<sub><25</sub> products and being enriched in multibranched isomers) is a different product compared to the prior art products (Schulze-Trautmann, paragraph 0095).

10. In order to demonstrate that both the process and the product as claimed is novel and non-obvious, in the following paragraphs are given:

- (i) the mechanistic background of hydroisomerization of long-chain n-paraffins over bifunctional catalysts,
- (ii) the analytical characterization of wax products obtained by catalytic experiments (which were conducted under my immediate supervision),
- (iii) the specific location of the hydrogenation metal component as described in the claims and the specification of Schulze-Trautmann within the binder-containing zeolite catalyst, and
- (iv) the effect of the metal-free zeolite component as described in the claims and the specification of Schulze-Trautmann on the enrichment of multibranched isomers within the microcrystalline paraffin.

11. Hydroisomerization and hydrocracking of n-paraffins is achieved on bifunctional catalysts. According to the bifunctional reaction scheme (original reports from Weisz [Adv. Catal. 13 (1962) 137-190] and Coonradt and Garwood [Ind. Eng. Chem. Proc. Design Dev. 3 (1964) 38-45]), the n-paraffin is dehydrogenated on the hydrogenation/dehydrogenation component and the corresponding n-olefin is protonated on the acid site. The alkylcarbenium ion formed upon protonation undergoes skeletal rearrangements and, eventually, cracking through  $\beta$ -scission.  $\beta$ -scission becomes more and more favorable as

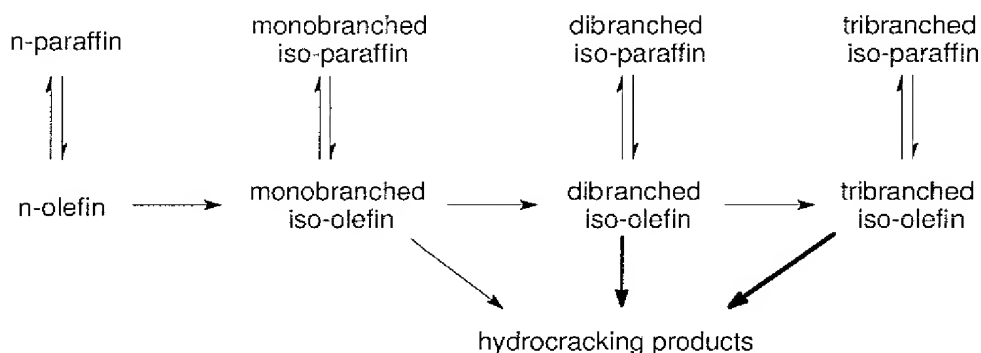


Fig. 1: Reaction pathways of hydroisomerization  
the branching degree of the carbon chain increases (indicated by bold arrows in Fig. 1).  
The desired iso-paraffins are formed by hydrogenation of the corresponding iso-olefins.

12. High temperature/resolution gas chromatography (see Schulze-Trautmann, 0072) is an appropriate analytic technique to reveal the detailed composition of mixtures of long chain paraffins in comparison to the previous characterization of the prior art paraffin waxes, e.g. by pour and melting point, boiling range, and needle penetration (see Bertaux, Eilers, and Wittenbrink).

Examples of the preferred feed of the hydroisomerization (synthesis product of the Fischer-Tropsch process) and the hydroisomerized product are given in Fig. 2. A more detailed composition regarding the single and multibranched isomers can be obtained by GC-MS analysis (Fig. 3). The peaks eluted between two n-paraffins, i.e.  $C_{i-1}$  and  $C_i$ , are ascribed to single and multi-branched  $C_i$  isomers (irrespective the findings for the hydroisomerisation of individual n- $C_i$  paraffins that some highly multibranched iso- $C_i$  are

eluted earlier than the corresponding  $n\text{-C}_{i-1}$  paraffin). The retention times of multibranched isomers are typically shorter than those of single branched isomers (see also Fig. 5). In general, the contents of iso-paraffins were determined by the summation of all peaks eluted between two  $n$ -paraffin peaks.

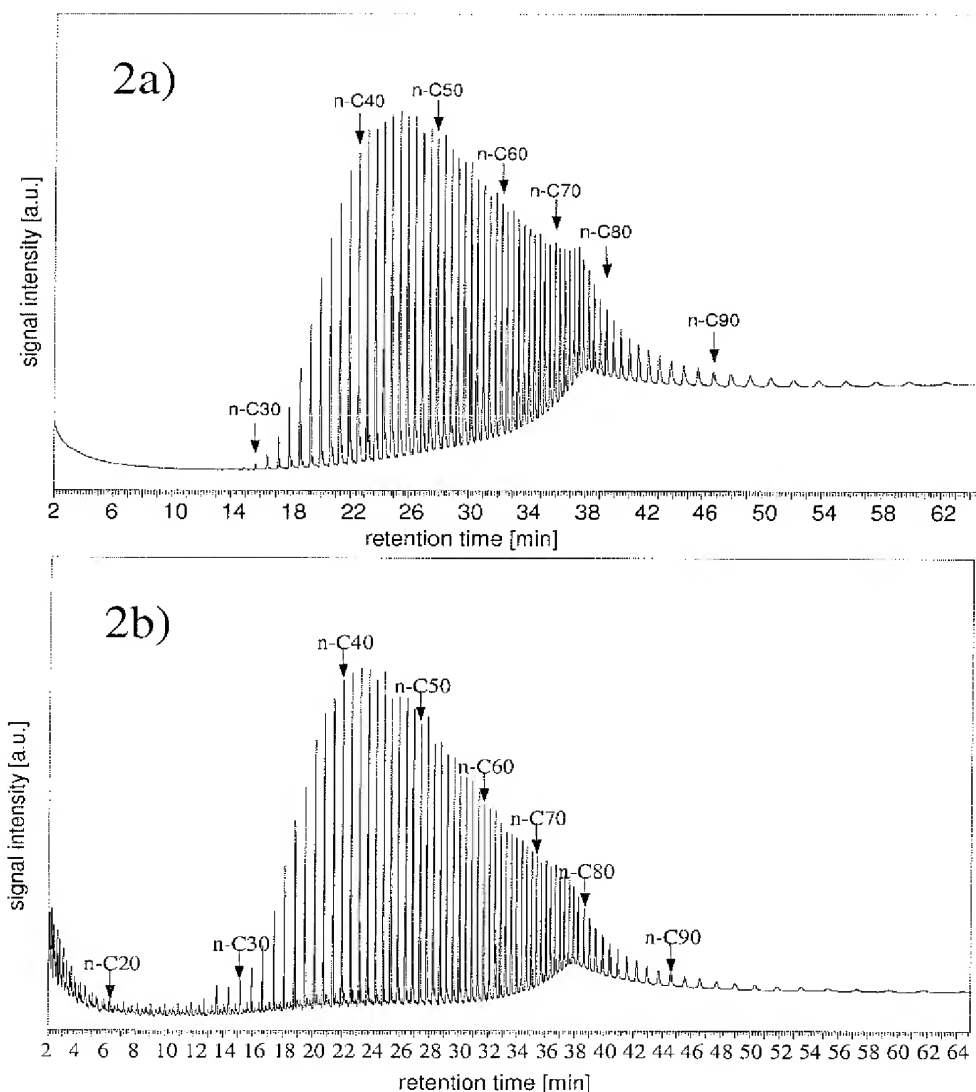


Fig. 2: High-temperature gas chromatograms of (2a) the preferred feed and (2b) the inventive microcrystalline paraffin.

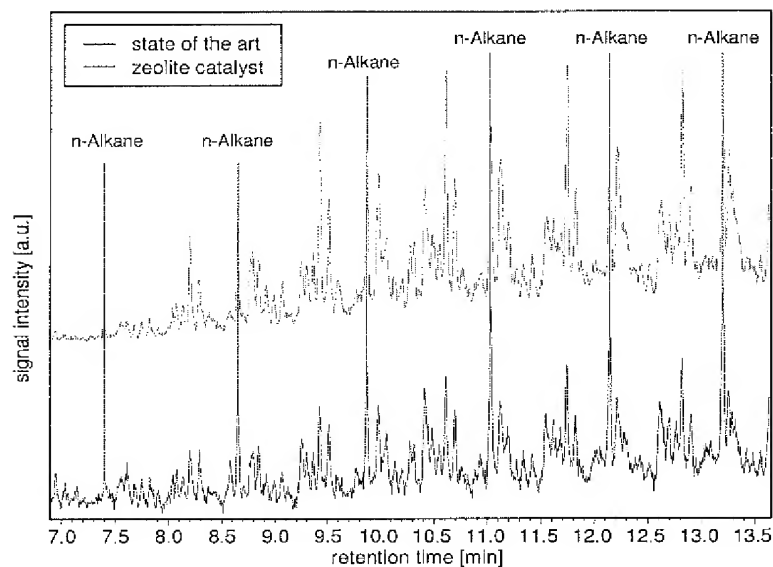


Fig. 3: GC-MS chromatograms of the inventive microcrystalline paraffin converted over the inventive zeolite catalyst and the wax products obtained on a prior art catalyst (Co-Mo/Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>).

13. The application of <sup>13</sup>C NMR spectroscopy (Hoek, 0019) indeed allows the determination of methyl-, ethyl-, and propyl-branching, but under the assumption that each molecule does not have more than one branch (which is an unrealistic precondition as later shown in Fig. 5). Obviously, pour and melting point, boiling range, and needle penetration of these wax mixtures can give only rough estimates of the composition (Bertaux, Eilers, Wittenbrink). For example, similar values of needle penetration can be obtained by a regular n-paraffin isomerisation conversion as well as by a low n-paraffin isomerisation conversion accompanied with enhanced cracking. Therefore, the position taken by the Examiner that Hoek (US2004/0199040) discloses a solid wax product which anticipates the inventive microcrystalline paraffin has respectfully to be rejected. The reasons for my opinion are set forth in the following paragraphs.

14. As visualized by Fig. 1, the hydroisomerized products have to be regarded as different mixtures of n- and iso-paraffins including single and multi-branched isomers and cannot be seen as one solid product prepared by catalytic hydroisomerization of FT paraffins irrespective the process/catalyst applied. The inventive microcrystalline paraffin as well as the inventive zeolite catalyst are mutual related. The physico-mechanical (and chemical) properties of the hydroisomerized FT waxes are controlled by the content of n- and iso-paraffins as well as the distribution of single and multi-branched isomers. Obviously, the content and the distribution of isomers are effectively influenced by the catalyst and the reaction conditions applied.

Compared to the prior art of microcrystalline paraffins, the inventive microcrystalline paraffin is characterized by

- (i) a reduced content of hydrocracked  $C_{<25}$  products,
- (ii) high iso-to-n paraffin ratios, and
- (ii) an enrichment in multibranched isomers.

15. In detail, the multibranched isomers as mentioned in Fig. 1 can be easily formed, but the formation of these bulky isomers is rapidly followed by cracking (in particular at higher reaction temperatures). If high yields of skeletal isomers have to be achieved, minimization of the hydrocracking reaction is mandatory. Hence, zeolites are good candidates for formulating hydroisomerization catalysts with low hydrocracking tendency because their shape selectivity reduces the formation of heavily multibranched isomers due to their pore sizes of molecular dimensions. Note, the beta zeolite applied to the invention (Schulze-Trautmann, 0049) has pores from about 0.5 to about 0.8 nm, compared to the pore diameters of silica-alumina (Hoek, 0012) in the range of 3.75 to 10 nm. In addition to steric restrictions preferring terminal branching, zeolites show, in general, strong acid sites and, thereby, rather low reaction temperatures (compared to amorphous silica-alumina catalysts), likewise reducing the tendency to hydrocracking. Therefore, non-zeolitic hydroisomerization catalysts (typically operating at higher reaction temperatures, e.g. 325 °C (Hoek, 0023) vs. 220°C (Schulze-Trautmann, 0098)) are, in general, causing a higher degree of cracking to low-molecular, liquid products

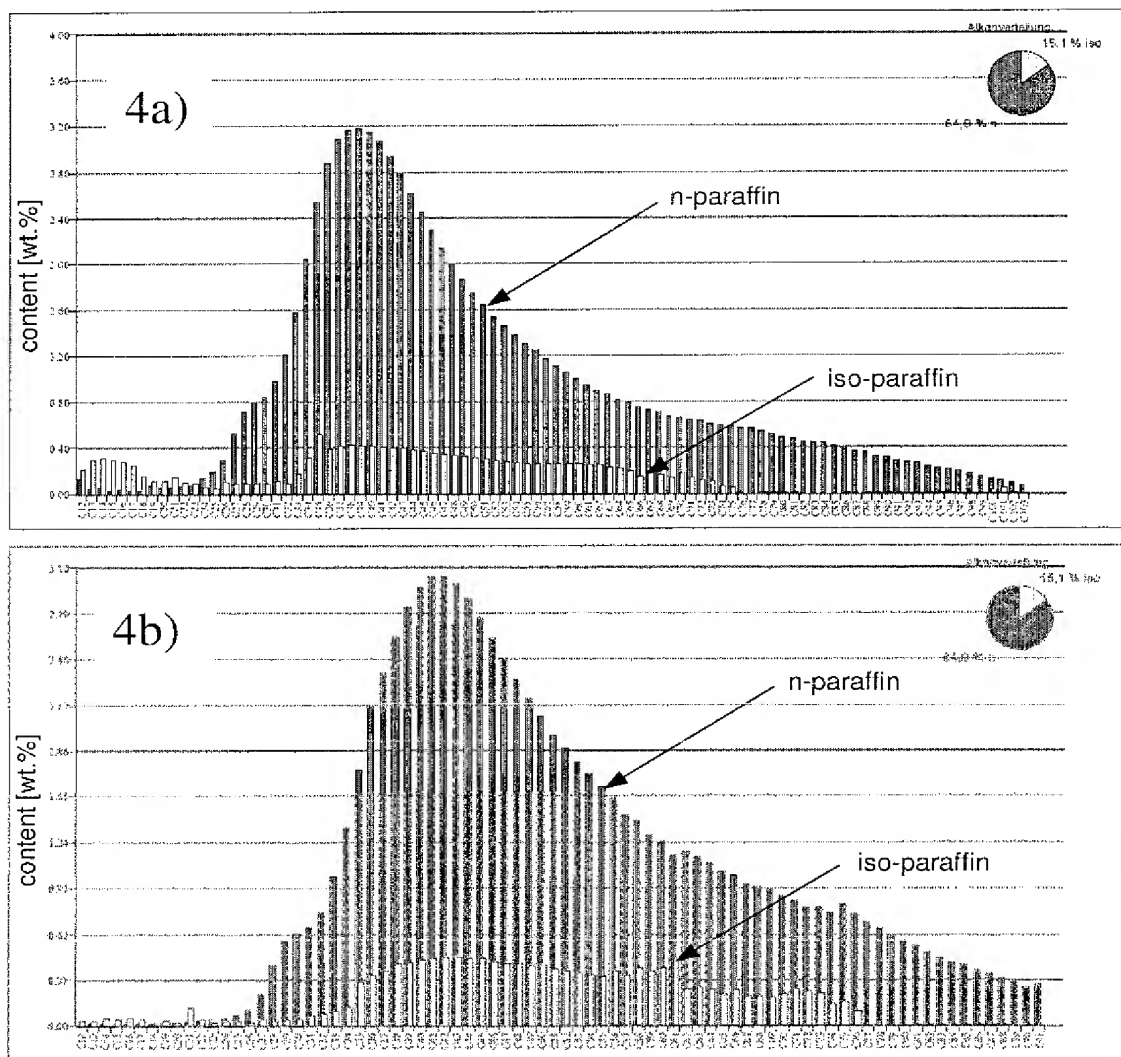


Fig. 4: Distribution of n- and iso-paraffins obtained on (2a) a prior art catalyst (Co-Mo/ $\text{Al}_2\text{O}_3$ - $\text{SiO}_2$ , 300°C) and (2b) the inventive zeolite catalyst (Pt/beta zeolite, 240 °C) for a similar ratio of iso/n-paraffins.

(indicated by the contents of  $\text{C}_{<25}$  paraffins in Fig. 4) at a similar degree of n-paraffin conversion. Obviously, the inventive microcrystalline paraffin features a different paraffinic composition compared to the prior art products, although the feed material for the production of all these microcrystalline paraffins is from a FT synthesis process.



16. For proper functioning of bifunctional catalysts, the hydrogenation/dehydrogenation sites control the concentration of olefinic intermediates, as well as the extent of multibranching. Obviously, low concentrations of olefinic intermediates result in low yields of iso-paraffins and too high concentrations may lead to enhanced hydrocracking. At reaction temperatures of about 240 °C, i.e. low reaction temperatures compared to amorphous silica-alumina catalysts of >300 °C, the extent of hydrocracking can be tolerated (Schulze-Trautmann, 0099) and there is still room for optimization and improvement. It has unexpectedly been found that higher contents of single- and multibranched isoparaffins can be effectively obtained by employing a bifunctional zeolite catalyst on which the hydrogenation/dehydrogenation sites are separated from the acid (zeolitic) sites. Therefore, the position No. 15 taken by the Examiner that "Hoek discloses the use of any suitable *amorphous* silica-alumina carrier (e.g. a *zeolite*)" as a support for a metal of transition group VIII for a hydroisomerization process which anticipates the inventive zeolite catalyst is not supported (it should kindly to point out that zeolites are not amorphous materials but crystals). Furthermore, Wittenbrink (WO 01/74969 A2, claim 3) explicitly claims that the metal components of the hydroisomerization catalyst are supported on an acidic support.

17. Because the zeolite crystals do not contain any hydrogenation metal component (Schulze-Trautmann, 0047 and 0055), the residence time of olefinic intermediates on the acid sites is effectively prolonged (compared to the prior art catalysts where the hydrogenation/dehydrogenation components are located in the immediate vicinity of the acidic sites) and, thereby, the probability of the formation of di- and tribranched isomers is increased resulting in both a higher iso/n paraffin ratio and a significantly higher content of multibranched isoparaffins compared to the prior art processes at a similar n-paraffin conversion (Fig. 5). These multibranched (trimethyl) isomers are not subject to enhanced hydrocracking due to the significantly lower reaction temperature of the inventive zeolite catalysts (Schulze-Trautmann, 0098) compared the prior art processes.

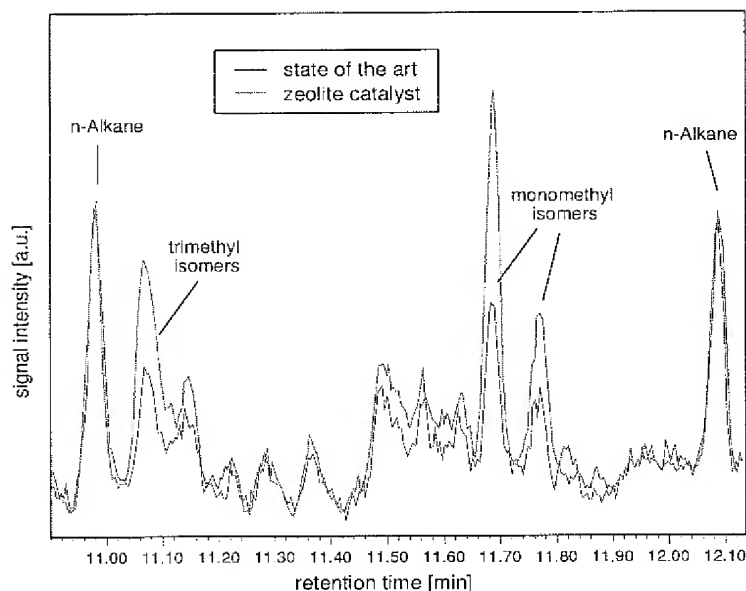


Fig. 5: GC-MS chromatograms of the inventive microcrystalline paraffin converted over the inventive zeolite catalyst and the wax products obtained on a prior art catalyst (Co-Mo/Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>).

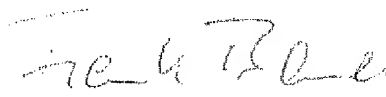
18. Regarding the individual paraffinic composition, the inventive microcrystalline paraffin shows considerably different ratios in n-/iso-paraffins and single/multibranched isomers compared to the prior art microcrystalline paraffins. Thus, the inventive microcrystalline paraffin has to be considered as a different product (having reduced contents of hydrocracked C<sub><25</sub> products and being enriched in multibranched isomers). In light of the prior art, the invention addresses the object of providing a novel microcrystalline paraffin and a catalytic process for preparing it.

19. It is my opinion that, given the surprising and unexpected results obtained by the application of bifunctional zeolite catalysts where the zeolite crystals do not contain any hydrogenation metal component and the hydrogenation/dehydrogenation sites are exclusively attached to the catalyst binder, the ordinary artisan would not have been able to predict or select these specific parameters, to produce the desired microcrystalline paraffin, having the claimed properties, because the prior art preparation of bifunctional zeolite catalysts consists of the impregnation or ion exchange of the hydrogenation

component onto the acidic support or zeolite (e.g., similar to that disclosed by Carati; column 4).

20. I declare further that all statements made in this Declaration of my own knowledge are true and that all statements made on information and belief are believed to be true and further, that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: March 30, 2010

A handwritten signature in dark ink, appearing to read "Frank Bauer", is written over a horizontal line.

Signed by: Frank Bauer

# **EXHIBIT 1**

## Curriculum Vitae Frank Bauer

Frank Bauer studied chemistry (Chemical engineering) at the University of Leipzig, Germany, where he also received his PhD in 1976. He joined the Academy of Science (ZfI Leipzig) as a research chemist and has been intensively involved in many applied research projects studying zeolite-catalyzed hydrocarbon reactions. In 1989, he got the habilitation degree at Academy of Science and has been Assistant Professor at University of Leipzig, since 1995. He moved to Leibniz-Institute of Surface Modification (IOM Leipzig) in 1998, where he continued his work on heterogeneous catalysis, isotope labelling, and nanoparticle applications.

Frank Bauer has published more than 100 technical papers, most of them related to zeolite catalysis and nanoparticle applications. He is co-author of the much-cited compendia "Handbook of Heterogeneous Catalysis", Wiley-VCH (Weinheim, 2008) and "Molecular Sieves-Science and Technology", Springer (Berlin, 2007). His recent research line is on the use of organosilanes as modifying agents for nanosized zeolites.